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The solar oxygen-isotopic composition: Predictions and implications for solar nebula processes

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Abstract–The outer layers of the Sun are thought to preserve the average isotopic and chemical composition of the solar system. The solar O-isotopic composition is essentially unmeasured, though models based on variations in meteoritic materials yield several predictions. These predictions are reviewed and possible variations on these predictions are explored. In particular, the two-component mixing model of Clayton and Mayeda (1984) (slightly revised here) predicts solar compositions to lie along an extension of the calcium-aluminum-rich inclusion (CAI) ^{16}O line between ($\delta^{18}O$, $\delta^{17}O$) = (16.4, 11.4)‰ and (12.3, 7.5)‰. Consideration of data from ordinary chondrites suggests that the range of predicted solar composition should extend to slightly lower $\delta^{18}O$ values. The predicted solar composition is critically sensitive to the solid/gas ratio in the meteorite-forming region, which is often considered to be significantly enriched over solar composition. A factor of two solid/gas enrichment raises the predicted solar ($\delta^{18}O$, $\delta^{17}O$) values along an extension of the CAI ^{16}O line to (33, 28)‰. The model is also sensitive to the nebular O gas phase. If conversion of most of the gaseous O from CO to H_2O occurred at relatively low temperatures and was incomplete at the time of CM aqueous alteration, the predicted nebular gas composition (and hence the solar composition) would be isotopically heavier along a slope 1/2 line. The likelihood of having a single solid nebular O component is discussed. A distribution of initial solid compositions along the CAI ^{16}O line (rather than simply as an end-member) would not significantly change the predictions above in at least one scenario. Even considering these variations within the mixing model, the predicted range of solar compositions is distinct from that expected if the meteoritic variations are due to non-mass-dependent fractionation. Thus, a measurement of the solar O composition to a precision of several permil would clearly distinguish between these theories and should clarify a number of other important issues regarding solar system formation.

INTRODUCTION

The average composition of the solar system is widely believed to be preserved in the outer layers of the Sun. Measured solar system bodies—including Earth, the Shergotty–Nakhla–Chassigny (SNC) and the howardite–eucrite–diogenite (HED) meteorites (Mars and Vesta respectively), and various meteorite and inclusion groupings—display fascinating detail in their O-isotopic composition (see Clayton, 1993, for review). However, the O-isotopic composition of the Sun, which contains ~99.8% of all solar system material, is largely unconstrained by observations. On the basis of CO absorption lines, Hall (1973) set an upper limit on the difference between the photospheric and terrestrial $^{18}O/^{16}O$ ratio at ±35%, and for $^{17}O/^{16}O$ a factor of 2.5. A more thorough high-resolution study of hundreds of CO lines by Harris *et al.* (1987) gave $^{16}O/^{18}O$ = 440 ± 50, or $\delta^{18}O \approx 140 \pm 130$‰ relative to standard mean ocean water (SMOW) ($^{16}O/^{18}O$ = 499). Collier *et al.* (1998) recently reported a solar wind $^{16}O/^{18}O$ ratio of 450 ± 130 ($\delta^{18}O \approx 110 \pm 290$‰) averaged over 17 months of spacecraft mass spectrometer data by *Wind*/MASS. [‰ = deviations per thousand (permil) relative to the standard. $(^{18}O/^{16}O)_{SMOW}$ = 0.0020052; Baertschi, 1976; Fahey *et al.*, 1987; Holmden *et al.*, 1997. $\delta^{17}O$ and $\delta^{18}O$ refer to permil deviations from SMOW.]

Figure 1 shows the isotopic structure so far observed within our solar system. The most notable feature in Fig. 1 is the trend extending from the main field of meteoritic and planetary compositions down to $\delta^{18}O \approx -60$‰, along which the Ca-Al-rich inclusions (CAIs) lie (*e.g.*, Clayton *et al.*, 1977). Russell *et al.* (1998) recently showed that there is a nearly parallel trend in the relatively rare ordinary chondrite Al-rich chondrules that extends down from ordinary chondrite bulk compositions. These lines have slopes of ~1 and can be interpreted in terms of enrichment or depletion of ^{16}O relative to the other O isotopes. We will refer to these trends as the "^{16}O lines." Variations along lines of slope ~0.5 can generally be understood in terms of mass-dependent isotope fractionation during both high and low-temperature processing in the solar system. Isotopic variations of bulk materials in the solar system are very small relative to the compositional range exhibited by the presolar sources of solar system O (*e.g.*, Nittler *et al.*, 1997; Timmes *et al.*, 1995). An important O measurement not shown in Fig. 1 is that of cometary water. Measurements by *Giotto* instruments gave a comet Halley water ice composition of $\delta^{18}O \approx 12 \pm 75$‰ (Eberhardt *et al.*, 1995; Balsiger *et al.*, 1995).

The measured O composition of the Sun presently has uncertainties extending well beyond the boundaries of Fig. 1. In principle, a more accurate solar composition could be estimated from the compositions of solar system solid materials if one could be confident of the processes involved in producing the isotopic variations in the solar nebula. No previous study has focused explicitly on theoretical predictions of the solar O-isotopic composition. Most studies have been concerned with explaining the O-isotopic variation in solar system bodies—though as discussed below, some have attempted to constrain the nebular gas composition, which has a strong relation to solar composition.

The hypothesis of a hot homogenized solar nebula, in which all solar system materials progressively condensed from the gas phase (*e.g.*, Cameron, 1962), has long been popular because of its relative success in explaining the bulk compositional properties of planets,

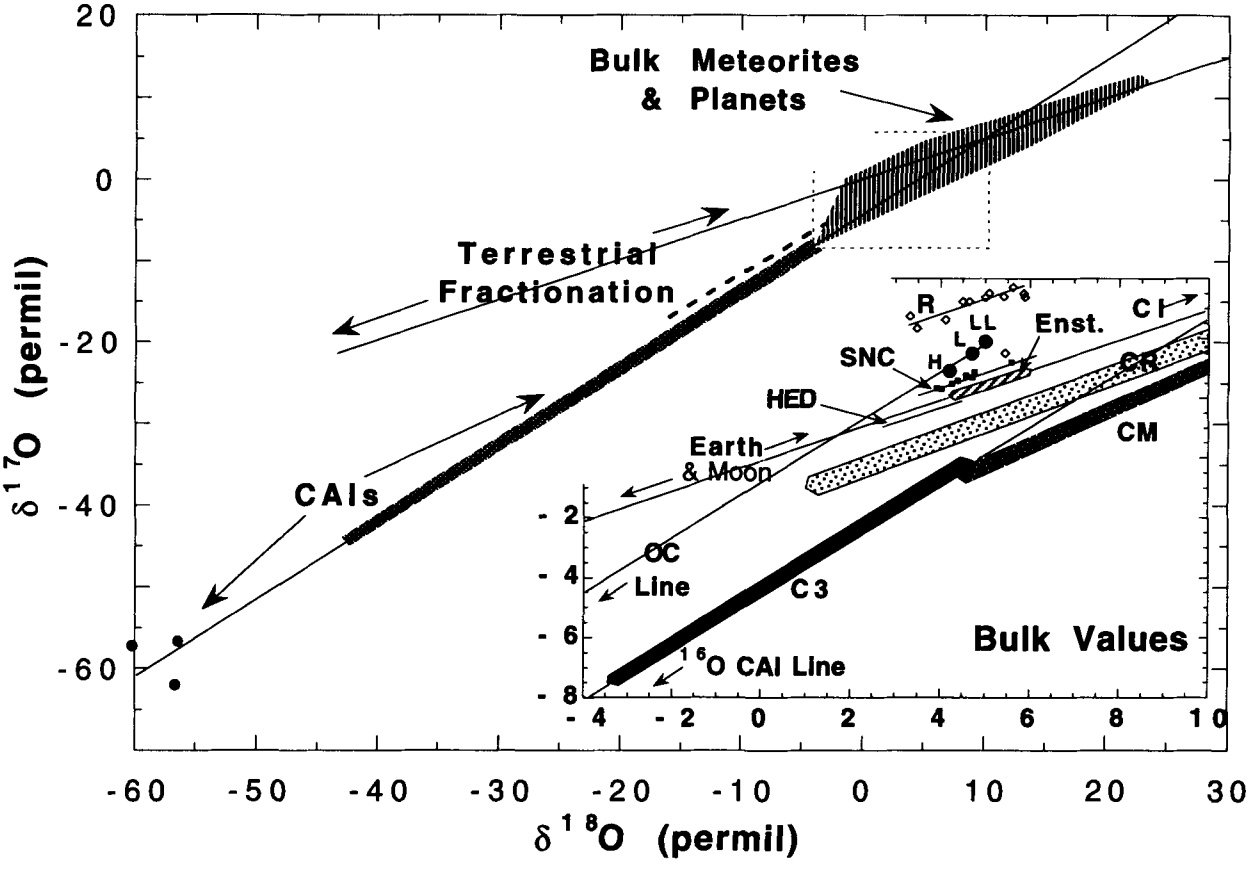

FIG. 1. Ranges of bulk O compositions for most of the meteorite classes (Clayton, 1993; Clayton and Mayeda, 1996; Russell *et al.*, 1998). The stippled area along the line labeled "CAIs" refers to mineral separates in Ca-Al-rich inclusions, whereas the points near the lower end of the line are selected extreme measurements made by ion probe (Fahey *et al.*, 1987; Virag *et al.*, 1991; Ireland *et al.*, 1992). Labels C3, CM, CR, CI refer to respective carbonaceous chondrite classes; H, L, and LL refer to the ordinary chondrite (OC) classes; R = R chondrites; Enst = Enstatites; SNC = Shergotty–Nakhla–Chassigny Martian subgroup; HED = howardite–eucrite–diogenite subgroup (= Vesta).

meteorites, and their constituents. Such a model would predict essentially identical solar and terrestrial isotopic ratios. However, the discovery of O-isotopic heterogeneity (Clayton *et al.*, 1973, 1977) and of large isotopic anomalies in Xe and Ne (*e.g.*, Reynolds and Turner, 1964; Black and Pepin, 1969; Lewis *et al.*, 1977) showed that presolar isotopic diversity in fact had survived the nebula phase to be incorporated into the meteorites. Although some aspects of the hot, homogeneous solar nebula envisioned by Cameron's (1962) model have fallen out of favor, it is an open question whether the composition of the Sun should be isotopically similar to the Earth. Wasson (1988) suggested that the solar nebula would have been hot enough to homogenize all isotopes in the inner solar system out to ~1 AU, implying that the Sun, Mercury, Venus, the Earth, and the Moon should all have essentially the same bulk O-isotopic compositions. Humayun and Clayton (1995) reached the same conclusion based on the homogeneity of solar system K isotopes in spite of the volatile-element depletions among terrestrial planets. As a variation on these suggestions, Choi and Wasson (1997) suggested that the O composition of the gas phase in the homogenized solar nebula might have changed with time enough to produce the entire meteoritic range of O compositions in solids formed from the evolving gas.

A longstanding explanation of the O-isotopic variation in the solar system within the context of an initially homogeneous solar nebula was described by Thiemens and Heidenreich (1983). They proposed that the CAI ^{16}O line and the meteorite data lying above the terrestrial fractionation line could be explained by non-mass-dependent fractionation processes operating on an initially homogeneous solar system composition. Photochemical non-mass-dependent isotope fractionation has been demonstrated to occur both in the laboratory and in the Earth's stratosphere and can produce reservoirs both enriched and depleted in ^{16}O (Thiemens and Heidenreich, 1983; Thiemens, 1996). The initial explanation for the non-mass-dependent fractionation was optical self shielding by the much more abundant $^{16}O^{16}O$ molecule (Thiemens and Heidenreich, 1983). But most recent work favors fractionation controlled by the greater stability of nonsymmetric molecules such as ^{16}O-C-^{17}O or ^{16}O-Si-^{18}O compared to the symmetric ^{16}O-C-^{16}O or ^{16}O-Si-^{16}O molecules (*e.g.*, Thiemens, 1996). Similar fractionations can also occur with AlO and FeO systems. The case for non-mass-dependent fractionation has been strengthened recently by new data from both Al-rich inclusions in ordinary chondrites (Russell *et al.*, 1998) and micro-analysis of CAI minerals (Young and Russell, 1998) that suggests that there exists an ^{16}O line with a slope of exactly 1.00, as required by the non-mass-dependent hypothesis. Previous data indicated a best single fit to the CAI data was a line of slope ~0.95. Thiemens has suggested (Thiemens, 1996; pers. comm., 1998) that if the non-mass-dependent fractionation model is correct, the solar composition should lie near the terrestrial fractionation line between the ordinary chondrite and CAI ^{16}O lines. While this prediction places more emphasis on CO-CO_2 chemistry, Nuth *et al.* (1998) focused

specifically on non-mass-dependent fractionation associated with SiO-SiO_2 chemistry. In his scenario, condensing silicates are expected to be isotopically heavy relative to the gas phase, which suggests that the solar composition will be near ($\delta^{18}O$, $\delta^{17}O$) ~ (–40, –40)‰ (Nuth *et al.*, 1998).

Clayton *et al.* (1973, 1977) suggested early on that the variations along the CAI ^{16}O line observed in components of carbonaceous chondrites (Fig. 1) might be due to variable gas–solid exchange between an ^{16}O-rich solid and a gaseous reservoir, each with its own nucleosynthetic history. The gaseous reservoir was assumed to be the solar nebular gas and the ^{16}O-rich solid reservoir was conceived to be a minor contributor to the solar system and so would not greatly perturb the inferred bulk solar composition. Oxygen is unique among major elements in the solar nebula in two ways: It is the only major element to show significant isotopic variations in bulk meteoritic materials, and it is the only major element to have significant reservoirs in both the nebular gas and solid phases. In this view, the isotopic variations are a direct result of the fact that O existed in both the gas and solid phases.

In order to explain the O-isotopic systematics of chondrules from carbonaceous, ordinary, and enstatite chondrites and the O compositions of the achondrites, Clayton and Mayeda (1985) postulated that solid reservoirs both more ^{16}O-rich and more ^{16}O-depleted than the terrestrial composition exchanged to varying degrees with a single gas reservoir having a composition essentially on the terrestrial fractionation line with ($\delta^{18}O$, $\delta^{17}O$) ~ (8.5, 4.2)‰. However, the bulk of the evidence seems to indicate in this context that the gaseous component of the nebula—and thus, the Sun—has a composition more ^{16}O-poor than the Earth (*e.g.*, Clayton and Mayeda, 1984; Rowe *et al.*, 1994; Choi *et al.*, 1998; Leshin *et al.*, 1997).

Under this assumption, Clayton and Mayeda (1984) expanded the original model of gas–solid exchange to include equilibration of partially exchanged solids with an aqueous fluid on the parent body to explain the O-isotopic systematics of Murchison (CM2) and other carbonaceous chondrites that have experienced pervasive aqueous alteration. This liquid reservoir was conceived to be derived from the gaseous reservoir that had previously exchanged with the ^{16}O-rich CAIs. Isotopic fractionations during the aqueous exchange were used to estimate water-to-rock ratios and the composition of the liquid reservoir. With only two initial components, the Clayton and Mayeda (1984) model is one of the simplest models for nebular mixing; and extended to the nebula as a whole, it is surely an oversimplification, especially as one is forced to postulate that all solar system material was initially one of two extreme compositions. Nevertheless, this model is the only one that permits clear investigation of the range of predicted solar compositions within the context of nebular solid–gas mixing.

In this paper, we will closely examine the Clayton and Mayeda (1984) model (hereafter, CM84) to see what limits can be placed on the solar composition. We will describe the model in the next section and compare and critique its utility in the section following. Our purpose is to see to what extent the O composition of the Sun can be constrained by the compositions of the meteorites, under the assumption that the O-isotopic variations in the solar system arise from incomplete homogenization of O from distinct nucleosynthetic sources, rather than from non-mass-dependent fractionation processes. In so doing, it should be possible to compare predictions of the solar composition between different scenarios of the nebular mixing model and the non-mass-dependent fractionation model. No previous study has focused directly on this issue.

In addition to the standard permil notation relative to SMOW (as defined above), two other notations will be used. One is

$$\Delta^{17}O = \delta^{17}O - 0.52\,(\delta^{18}O) \quad (1)$$

The $\Delta^{17}O$ gives the vertical distance from the terrestrial fractionation line (Fig. 1). The $\Delta^{17}O$ metric is useful to distinguish small differences that cannot be attributed to mass fractionation in the face of much larger mass fractionation effects, such as when comparing terrestrial and Martian samples. The other notation is the absolute difference from bulk Earth given by

$$[(\delta^{18}O - \delta^{18}O_{Earth})^2 + (\delta^{17}O - \delta^{17}O_{Earth})^2]^{1/2} \quad (2)$$

where ($\delta^{18}O_{Earth}$, $\delta^{17}O_{Earth}$) = (6, 3)‰, the average composition of terrestrial and lunar O (*e.g.*, Clayton and Mayeda, 1975). This metric gives an absolute difference in $\delta^{18}O$-, $\delta^{17}O$-space where mass fractionation is not a major issue. This is useful, for example, in estimating the isotopic precision required for a measurement of the solar composition to distinguish between the predictions of competing theories, assuming that the specific predictions and measurements are relatively immune to mass fractionation. The bulk Earth baseline is used not for ethnocentric reasons, but because one of the non-mass-dependent fractionation predictions is for solar composition to be nearly the same as bulk Earth. Thus, the absolute difference from bulk Earth conveniently approximates the absolute difference from one of the major solar predictions.

EXISTING TWO-COMPONENT MODEL PREDICTIONS

The CM84 Model

The CM84 model addressed Murchison (CM2) and, to a lesser extent, Orgueil (CI). The mineralogies of these meteorites reflect high degrees of aqueous alteration (McSween, 1979). The authors assumed that the solid precursors to the CI and CM2 chondrites had a composition near the ^{16}O-rich extreme of the CAI trend, whereas initial solar system gas had a composition somewhat more ^{16}O-poor than the most extreme observed carbonaceous component (Fig. 1). The composition of the gas was calculated from the equations and assumptions reviewed here. In the first step, the initial nebular compositions of solids, *S*, and gas, *G1*, were modified by partial isotopic exchange to produce the anhydrous silicates measured in Murchison, with an average value of ($\delta^{18}O$, $\delta^{17}O$) = (–4.2, –7.4)‰, point *A* in Fig. 2 (parentheses notation for $\delta^{18}O$, $\delta^{17}O$ will be used hereafter). This partial exchange is schematically illustrated by

$$S + G1 \Rightarrow A + G2 \quad (3)$$

Further modification of solids occurred by aqueous alteration with water (*L1*) condensed from the partially exchanged nebular gas source,

$$G2 \Rightarrow L1 \quad (4)$$

Equilibrium thermodynamics predicts that O in the gas at high temperature should be distributed between CO and H_2O; whereas at low temperature, the O should be almost entirely in the form of H_2O. The CM84 model assumed that gaseous O was essentially all in the form of H_2O. Aqueous alteration of solids (*A*) resulted in matrix phyllosilicates that in Murchison have a composition given by *M*. Calcite was precipitated with a measured composition *C*.

$$A + L1 \Rightarrow M + C + L2 \quad (5)$$

The relatively large fractionation between phyllosilicates (*M*) and calcite (*C*) in Murchison suggests a very low equilibration tem-

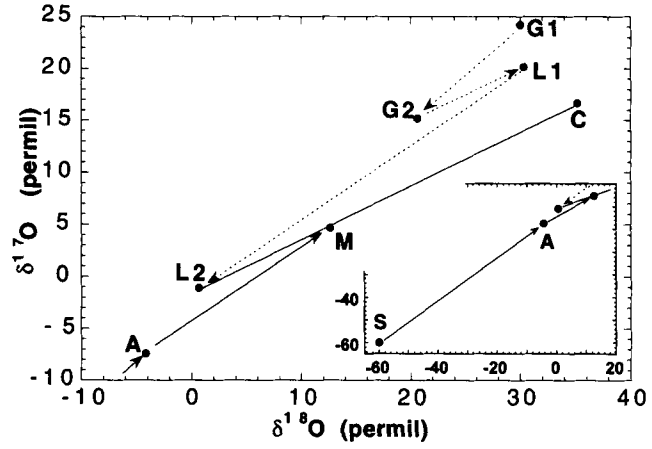

FIG. 2. The CM84 model for the isotopic history of Murchison. Initial nebular compositions of solids, *S*, and gas, *G1*, partially exchanged to produce compositions *A* and *G2*. Interaction with condensed water, *L1*, produced compositions *M* and *L2*. Calcite of composition *C* was precipitated. Composition *L2* was estimated by plausible fractionation from *C*. The compositions of *L1*, *G2*, and *G1* were then calculated from *S*, *A*, *M*, and *L2*. Compositions are given in Tables 1 and 2.

perature, clearly less than ~50 °C. The CM84 model estimated the alteration temperature in Murchison at between 0 and 20 °C. The final isotopic composition of the water, *L2*, is fixed by the calcite–water fractionation (O'Neil *et al.*, 1969). Using the *L2* point and mass balance considerations, the initial nebular gas composition, *G1*, was then determined as follows: The length of *L1–L2* is proportional to *A–M*, related by the ratio, *f*, of O in the water to O in the rock. The line *G2–L1* represents the fractionation during condensation of water from the gas phase, where a simple batch condensation was assumed. The equations thus derived are

$$\delta^{18}O_{G2} = \delta^{18}O_{L2} + (\delta^{18}O_M - \delta^{18}O_A)/f - 11.40\,(1 - f \times R) \quad (6)$$

$$\delta^{17}O_{G2} = \delta^{17}O_{L2} + (\delta^{17}O_M - \delta^{17}O_A)/f - 5.92\,(1 - f \times R) \quad (7)$$

where the first portion of the equations determines *L2–L1* and the second part is the condensation step, and where *R* is the ratio of O from solid *vs.* gas components in the nebula. The equations are derived in greater detail in the original paper (CM84). Errors from using the linear mass fractionation approximation (slope = 0.52) rather than the exponential curve are ≤0.1‰, which is similar to in CM84. The length of *G1–G2* is proportional to *S–A* and *R*. Points *G2* and *G1* are displaced slightly from an upward extension of the CAI ^{16}O line. This displacement is due to solid/gas fractionation, which is insensitive to the relative amounts of CO and H_2O(g) and to temperature over hundreds of degrees (Onuma *et al.*, 1972). The CM84 model used a gas–solid partial exchange temperature of 1800 °C and a CO/H_2O ratio of 9, both adopted here. The resulting line along which *G1* and *G2* lie is given by CM84 as

$$\delta^{17}O = 0.95\,(\delta^{18}O) - 4.34 \quad (8)$$

Thus, the system is one of three equations and three unknowns (f, $\delta^{18}O$, $\delta^{17}O$), where f is the water/rock ratio.

Table 1 reproduces the CM84 points shown in Fig. 2, but with the following minor changes: (1) More recent observations, particularly by ion probe (*e.g.*, Fahey *et al.*, 1987; Virag *et al.*, 1991; Ireland *et al.*, 1992; McKeegan *et al.*, 1998), suggest a more ^{16}O-rich endmember for the solid component *S*, approximately ($\delta^{18}O$, $\delta^{17}O$) = (–60, –60)‰. It is clear that compositions more extreme than

TABLE 1. Input parameters for calculation of solar nebula gas. Composition based on Murchison phases, after CM84.

Phase	Symbol	$\delta^{18}O$ (‰)	$\delta^{17}O$ (‰)
Initial solids	*S*	–60	–61.3
Mean anhydrous solids	*A*	+4.20	–7.42
Matrix	*M*	+12.61	+4.69
Calcite	*C*	+35.14	+16.67
Final liquid (0° C equil.)	*L2*	+0.60	–1.12
Final liquid (20° C)	*L2*	+5.6	+1.5
Nebular O solid/gas ratio	*R*	18/82	

(–40, –40)‰—used by CM84—are much less abundant. However, the two-component model forces one to use the most extreme composition. The issue of whether the initial solids were distributed along the ^{16}O lines rather than as a simple endmember will be discussed later. (2) The assumed ratio of initial nebular O in the solid component to the gas component has been calculated from Anders and Grevesse (1989), giving R = 18/82 rather than the 21/79 used by CM84. The results are shown in Table 2 for Murchison water–calcite equilibration temperatures of 0 and 20 °C.

Figure 3 shows the predicted *G1* and *G2* composition ranges. From these, a simplistic prediction of the solar isotopic composition can be made assuming the initial *S* and *G1* compositions apply to the nebula as a whole, and not just in the CI-CM meteorite forming region. Combining these components in the proportions indicated by the solar elemental composition, *R*, gives a predicted solar compositional range between ($\delta^{18}O$, $\delta^{17}O$) = (16.4, 11.4) and (12.3, 7.5)‰, shown in Fig. 4. These values are ~1‰ higher than would be obtained using the CM84 values for *R* and *S* instead of those in Table 1.

Rowe *et al.* (1994), and subsequently Leshin *et al.* (1997), extended the CM84 model to new data obtained from CI chondrites. Although CM84 estimated an equilibration temperature of 150 °C for CI carbonates, a lower temperature was required by Leshin *et al.* (1997) to satisfy the equations. In addition, the existence of magnetites unequilibrated with the phyllosilicates—and with $\Delta^{17}O$ as low as +1.6‰—suggests that *L2* should be at least this low, implying a temperature no higher than 50 °C for equilibration. Solving the equations for this temperature gives f = 0.67, which is still higher than the 0 °C Murchison case, as would be expected. With the initial assumptions for *S* and *R* in Table 1, using the Leshin *et al.* (1997) value of f results in *G1* = (28.3, 22.9)‰ and a predicted solar composition of (12.4, 7.8)‰, which is almost identical to the 20 °C Murchison case (Table 2, Fig. 3).

TABLE 2. Compositions calculated from Table 1 parameters.

		0° C*		20° C*	
Phase	Symbol	$\delta^{18}O$	$\delta^{17}O$	$\delta^{18}O$	$\delta^{17}O$
Initial liquid	*L1*†	31.0	20.7	25.2	15.6
Final gas	*G2*	20.9	15.5	16.0	10.8
Initial gas	*G1*	33.2	27.4	28.2	22.7
Moles O in liquid / Moles O in solid	f	0.554		0.856	
Predicted solar composition		16.4	11.4	12.3	7.5

*Assumed aqueous alteration temperatures in Murchison.
†$\delta^{18}O$, $\delta^{17}O$ values in permil; f is given as a ratio.

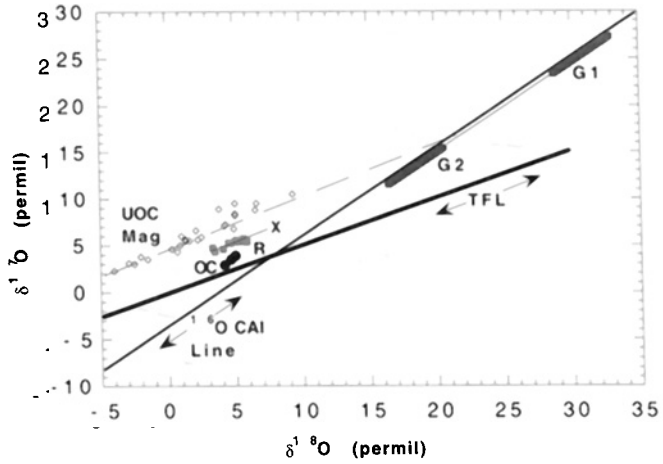

FIG. 3. Compositions of various predicted solar nebula gas components. The range of initial nebular gas, *G1*, in the revised CM84 model, is at the top right. The composition range after partial exchange, *G2*, is lower along the extension of the ^{16}O line defined by CAIs. The ranges *G1* and *G2* are slightly offset from the CAI ^{16}O line due to gas–solid fractionation (Onuma *et al.*, 1972). The upper ends of the *G1* and *G2* ranges correspond to the CM 0° C case; the lower ends represent nearly coincident points for the CM 20° C and CI cases. The upper end of the dashed line through Semarkona and Ngawi magnetites (UOC Mag) indicates the nebular composition suggested as the reservoir for oxidation of these minerals (Choi *et al.*, 1998). The "×" above and to the left of the R chondrites indicates the composition of gas in equilibrium with the R chondrites, which have the highest bulk meteorite Δ^{17}O (Clayton, 1993). TFL = terrestrial fractionation line; OC = ordinary chondrites.

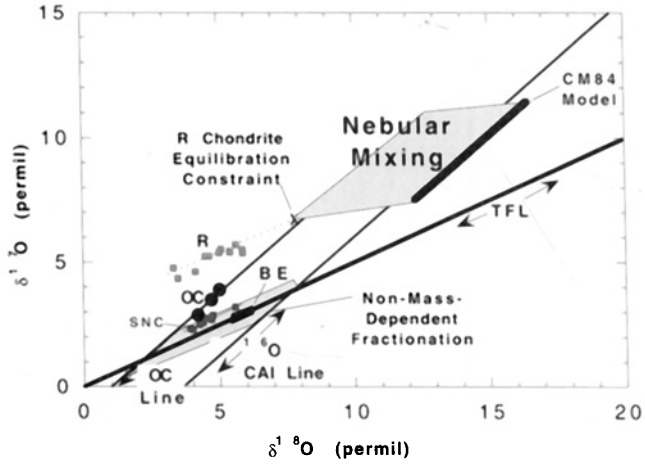

FIG. 4. Predicted solar compositions. The CM84 model predicts a composition range shown by the heavy line to the right of the ^{16}O line and above the terrestrial fractionation line. The "×" above and to the right of the R chondrites indicates the solar composition expected if nebular gas was completely equilibrated with these solids (Clayton, 1993). Due to the inclusion of solids, the "×" in this figure is slightly to the left of the gas reservoir indicated ("×") in Fig. 3. Labeled data points are similar to Fig. 1. As discussed in the text, "BE" = bulk Earth. The region between the extension of the ordinary chondrite ^{16}O line and the CM84 model results is shaded to indicate the range of likely solar compositions within the context of nebular solid/gas mixing. The shaded region along the terrestrial fractionation line (TFL) between the CAI and ordinary chondrite ^{16}O lines indicates the likely solar composition predicted for the case of non-mass-dependent fractionation (Thiemens, 1996), though in some scenarios the non-mass-dependent fractionation solar prediction could be much more ^{16}O-enriched (Nuth *et al.*, 1998).

Other Constraints

There are several other relevant constraints on the solar O-isotopic composition in the context of two-component solid–gas models. One such constraint is given by Choi *et al.* (1998), who measured O in unequilibrated ordinary chondrite magnetite grains (Fig. 3). These have been interpreted as low-temperature products of reaction of metal with nebular gas or water-rich vapor, or more likely, water condensed from nebular gas. In any case, these may represent an independent tie point to gaseous nebular O. On the basis of the O composition of the magnetites, Choi *et al.* (1998) suggested a nebular component approximately (21, 17.5)‰, intermediate between *G1* and *G2* compositions (Table 2). Most of the high-temperature gas–solid exchange in the nebula probably already occurred prior to magnetite formation, as its formation is a relatively low temperature process, which suggests that the magnetite reflects the final nebular gas composition, *G2*. By comparison with the CM84 model results, this infers a slightly higher final nebular gas composition.

Another constraint comes from direct gas–solid equilibration, thought to have been the predominant equilibration mechanism for ordinary chondrites in which aqueous alteration did not occur. Solid/gas equilibrium fractionation is relatively insensitive to temperature over the ~1200–1800 °C range in which it is effective and leaves the gas enriched in δ^{18}O ~2‰, relative to the final solid composition (Onuma *et al.*, 1972; Yu *et al.*, 1995). Assuming all solid materials have undergone some degree of gas–solid isotopic exchange, the resulting solar composition is constrained to be more ^{16}O-depleted than the most ^{16}O-depleted solid materials in Fig. 1. The meteorites farthest above the terrestrial fractionation line are the R chondrites (Weisberg *et al.*, 1991; Clayton, 1993), with an extreme value of (δ^{18}O, δ^{17}O) ~ (6, 6)‰ (Fig. 3). If this composition represents completely equilibrated solids, the final gas composition would be approximately (8, 7)‰ (Clayton, 1993). Considering both (8, 7)‰ gas and (6, 6)‰ solid inputs to the Sun, using ratio *R* in Table 2, the lowest solar (δ^{18}O, δ^{17}O) values permitted by the R chondrites is approximately (7.6, 6.8)‰. This is referred to in Fig. 4 as the "R Chondrite Equilibration Constraint." Note that this constraint permits a minimum solar (δ^{18}O, δ^{17}O) that is lower than the CM84 model predictions (Table 2)—particularly for δ^{18}O—and lies near an extension of the ordinary chondrite ^{16}O line.

DISCUSSION

Figure 4 summarizes the predictions of solar composition from the above studies. All of the CM84 model results are constrained to lie slightly to the right of the CAI ^{16}O line, which is significantly above the terrestrial fractionation line. If this exercise could be performed using ordinary chondrites and their corresponding ^{16}O line, the results would no doubt lie farther to the left, as indicated by the leftward extent of the shaded region. The predicted CM84 model solar composition has a Δ^{17}O value between 1.1 and 2.9‰, but it is also enriched in δ^{18}O relative to Earth and Mars. In terms of the absolute difference from bulk Earth, as defined in Eq. (2), the solar prediction is in the range of 7.7–13.4‰. A solar composition in this range would clearly indicate that isotopic homogenization in the solar nebula did not occur out to 1 AU. The CM84 composition is distinct from the non-mass-dependent fractionation predictions, near the intersection of the ^{16}O lines and the terrestrial fractionation line (Thiemens, 1996; pers. comm.), or below (Nuth *et al.*, 1998). At the closest, the CM84 and non-mass-dependent fractionation predictions are ~6‰ apart in absolute terms. Figure 4 shows the ranges of expected solar values as shaded regions. The shaded region for nebular mixing includes consideration of ordinary chondrite data

as well as the CM84 prediction. Considering this wider range, the minimum difference between nebular mixing and non-mass-dependent fractionation predictions is shown as ~3‰. The nebular mixing range is also distinct from that expected in the case of temporal evolution of the solar nebula from light to heavy gas (*e.g.*, Choi and Wasson, 1997). The exact composition in that case depends on when in the evolution process the Sun accreted most of its O, though Wasson (pers. comm., 1998) suggests the solar composition would most likely be below the terrestrial fractionation line.

Thus we have distinct predictions for the solar composition based on the nebular processes giving rise to the O-isotopic variations. However, it is necessary to go back and examine some of the assumptions made in the simplistic two-component model described above.

Solid/Gas Ratio

Our extension of the CM84 model assumes that the relative proportions of gas and solid components in the meteorite-forming region were the same as that accreted to the Sun. This is a major assumption, as the meteorite-forming region is often thought to have been significantly enriched in solids. To account for chondrules as liquid droplets, many studies (*e.g.*, Fegley and Palme, 1985; Wood and Hashimoto, 1993; Yoneda and Grossman, 1995) have considered equilibrium mineral assemblages in nebular environments with solid/gas as high as $R \sim 10^7$ times solar composition. The Table 1 parameters do not work for enrichments $R > 3.4$ times solar composition. Higher enrichments would require either moving *M* closer to the CAI ^{16}O line or moving *L2* farther to the left of the line (Fig. 2). However, *M* is fixed by the data and, for 0 °C equilibration temperature, *L2* is as far to the left as allowed by the calcite (Table 1). Figure 5 shows the trend from 0 to 200% solid/gas enrichments relative to the CM84 0 °C case (Tables 1 and 2). It is clear that a small enrichment of the dust/gas ratio in the meteorite region has a strong effect on the predicted solar composition. The predicted solar composition increases in both the absolute difference from terrestrial (to 37‰ at 200% solid/gas enrichment) and in $\Delta^{17}O$ (to 10.5‰ for the same solid/gas enrichment). Even if the CM84 model were manipulated to work to higher enrichment factors, the astronomical observations mentioned in the opening paragraph, as well as comet

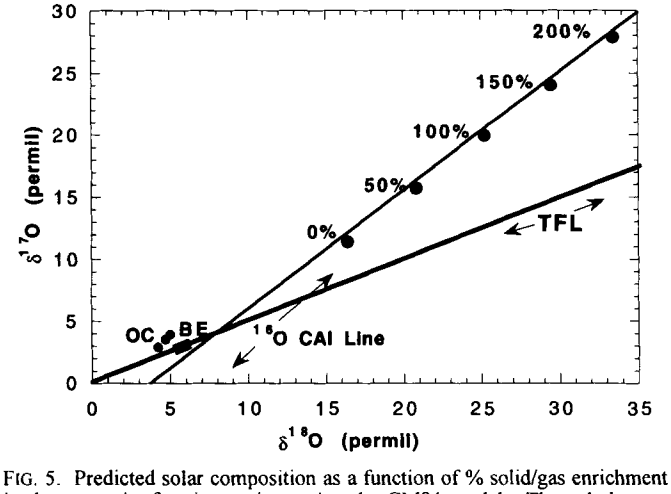

FIG. 5. Predicted solar composition as a function of % solid/gas enrichment in the meteorite-forming region, using the CM84 model. The nebular gas reservoir composition G1 moves up along the line, reaching $\delta^{18}O = 54$ by 200% solid/gas enrichment, and resulting in the solar composition shown. Mean H, L, and LL ordinary chondrite (OC) compositions and bulk Earth (BE) are shown for reference.

Halley H_2O observations, if representative of the nebular gas, would limit predicted solid/gas enrichments to below a factor of ~10. However, these observations may not be in direct conflict with theories invoking high *R* in the meteorite-forming region, as the sequence of events is important. Even if ratio *R* in the chondrule-forming region is high at any given time, gas exchange over a prolonged period, during which gas cycled in and out of this region, will reduce the effective solid/gas ratio in terms of the effect on O isotopes. Thus, the effective solid/gas fractionation might be compatible with the CM84 model even if the local solid/gas ratio is somewhat higher. Other possible perturbations to the predicted solar O composition—based on differences in the elemental composition of the solar nebula relative to the present-day solar surface—are discussed in the appendix but are generally small in magnitude.

Nebular Oxygen Gas Phase

For a nebular gas at equilibrium, CO is the dominant O species at temperatures above ~750 K. At lower temperatures at equilibrium, CO reacts with H_2, leaving essentially all O bound as H_2O. The CM84 model assumes that O in the gas phase of the nebula was essentially all in the form of H_2O. The initial gas–solid partial exchange, Eq. (3), is regarded as being independent of the state of nebular O. The issue is whether nebular O was all in the form of H_2O(g) at the time liquid water condensed, Eq. (4). The condensation step is envisioned to occur in the relatively cool outer region where carbonaceous chondrites were formed. The bulk of the O at closer distances to the protoSun would be hotter and still in the gas phase. Conversion from CO to H_2O is kinetically inhibited (Lewis and Prinn, 1980), so that some O could still be CO at the time of H_2O condensation. One resulting possibility is that the nebular H_2O and CO were in isotopic equilibrium at high temperature; but at some point as the nebula cooled, a "freezing in" of the isotopic difference between CO and H_2O occurred. In this case, high-temperature partial exchange of solids with the gas, Eq. (3), would have taken place with both CO and H_2O, while the water alteration phase of the carbonaceous chondrites would occur with H_2O, whose isotopic composition reflected the freezing-in temperature of the nebular gas. Another possibility is that a freezing-in point was never reached: CO and H_2O were in isotopic communication at the H_2O condensation temperature, but at the time of water condensation for the carbonaceous chondrites, only a small fraction of the O had been converted to H_2O. Equilibrium fractionation between CO(g) and H_2O(g) causes the H_2O to be isotopically lighter. The strongest effect would be observed if the H_2O(g) in Eq. (4) was converted from CO(g) at the lowest temperatures. This would result in the following additional step in the sequence:

$$G2\,(\mathrm{CO}) \Rightarrow G2(\mathrm{H_2O}) \qquad (9)$$

which has a mass-dependent fractionation of $\delta^{18}O = -18$‰ at 500 K (Onuma *et al.*, 1972). Higher-temperature conversion or freeze-in would have a lesser effect (*e.g.*, –4.6‰ at 1000 K). When this step is figured into Eqs. (6) and (7), the calculated initial gas component *G1* is shifted up and to the right along a slope 1/2 line. For the 500 K conversion mentioned above, *G1* would become ($\delta^{18}O$, $\delta^{17}O$) = (51.2, 36.7)‰ and the predicted solar composition would become (31.2, 19.1)‰. The $\Delta^{17}O$ value does not increase.

Equation (9)—as well as Eqs. (3) and (4)—can result in temporal evolution of the nebular gas composition. Equation (3) will simply result in evolution parallel to the ^{16}O lines. On the other hand, Eqs. (4) and (9) will result in evolution along mass dependent fractionation

lines (*e.g.*, the absolute difference from bulk Earth will change, but the $\Delta^{17}O$ value will be unchanged relative to the original CM84 predictions). Because the Sun contains >99% of the solar system O, it is highly unlikely that removal of O from the nebular reservoir to form planetary bodies will have any measurable effect on the solar composition. In spite of this, the gas reservoir in the meteorite-forming region may have evolved with time independent of the proto-solar material. Equation (9) indicates that removal of O in the form of $H_2O(g)$ will result in progressively heavier O in the remaining reservoir, effectively causing the ^{16}O lines to evolve to the right. Condensation of liquid water from $H_2O(g)$ (Eq. (4)) would cause, in contrast, a slight evolution of the remaining gas to the left.

Additional Solid Components

The two-component O isotope model discussed to this point is obviously the simplest possible case. The greatest argument for only two components comes from the fact that the CAI compositions lie on a relatively tight line. However, it is more likely that solids were distributed along the ^{16}O lines, rather than simply as an endmember. It is clear, for example, that a much larger number of grains are found near $(\delta^{18}O, \delta^{17}O) \sim (-40, -40)$‰ than further down the line. Distribution along the line might be expected if the average composition of interstellar solids evolves with time due to preferential recent input of ^{17}O and ^{18}O (*e.g.*, Timmes *et al.*, 1995). Because an isotopic composition can only be preserved for an extended period in a solid, and because interstellar solids are repeatedly subjected to shocks that can evaporate them, refractory material inherited by the solar system would be, on average, both older and more ^{16}O-rich than the more volatile material. If this is the case, the ^{16}O lines could extend to much more ^{16}O-rich compositions, but the number of grains would decrease with increasing ^{16}O enrichment due to their greater average galactic residence time. The majority of solids might be expected to have compositions similar to that of the gas, which carries the integrated input from all galactic sources. The average solid composition would thus be isotopically heavier than assumed by the CM84 model, resulting in a somewhat less extreme nebular composition for the gas as well. However, it is very important to note that within this model, even if the average initial solid and gas compositions were less extreme than those in Tables 1 and 2, the predicted solar composition does not change significantly and remains distinct from other predictions. As an example, if 70% of the solids had the same composition as the gas, with the remainder at the same composition as *S* in Table 1—assuming *S* was still the precursor for the carbonaceous chondrites—the predicted solar composition would be ~4.0‰ heavier in both $\delta^{18}O$ and $\delta^{17}O$ relative to the predicted range in Table 2. Other scenarios can be envisioned, but as this example shows, the CM84 model is surprisingly insensitive to additional solid components along the ^{16}O lines, and the uniqueness of the solar mixing model prediction, relative to other predictions, still holds.

CONCLUSIONS

Table 3 summarizes the models and observations discussed in this paper. As shown by the first two rows, the solar O-isotopic composition is relatively unconstrained by observations to date of either the Sun itself or by the solar wind emanating from it. The remainder of the table addresses the models discussed above, all of which assume that gases and solids are intimately linked in some way. If there were no gas–solid interaction in the meteorite-forming region, the solar composition would only be constrained by the $^{18}O/^{16}O$ observations of the first two rows of the table, limiting the difference between solar and terrestrial $\delta^{18}O$ to less than several hundred permil. On the other extreme, if solids and gases completely homogenized in the inner solar system to 1 AU, the solar O-isotopic composition must be essentially identical to terrestrial. Only the nebular gas–solid mixing models predict solar compositions significantly above the terrestrial fractionation line (TFL). If the CM84 mixing model is valid for O isotopes on a solar nebula-wide scale, the solar composition is most likely between $(\delta^{18}O, \delta^{17}O) = (12.3, 7.5)$‰ and (16.4, 11.4)‰, which is clearly distinct from the terrestrial fractionation line. Consideration of the ^{16}O line passing through the ordinary chondrites would allow somewhat lower $\delta^{18}O$ compositions (in line with the R chondrite equilibrium constraint) as shown by the shaded region in Fig. 4. The CM84 model results have a $\Delta^{17}O$ range of 1.1–2.9‰. However, the absolute permil difference from the bulk Earth composition, which is a better measure of the permil resolution needed to distinguish between solar predictions, is much greater at 7.7–13.4‰, as shown in the last column of Table 3. The main competing O isotope theory to solid–gas mixing—that of non-mass-dependent fractionation—predicts a distinct solar composition near the intersection of the TFL and the ^{16}O lines, that is, nearly identical to the bulk Earth (*e.g.*, Thiemens,

TABLE 3. Summary of solar O composition observations and predictions.

Model/constraint	Reference	Composition			Abs. diff. rel. bulk Earth (‰)*
		$\delta^{18}O$ (‰)	$\delta^{17}O$ (‰)	$\Delta^{17}O$ (‰)	
Solar CO line measurement	Harris *et al.* (1998)	140 ± 130	–	Unconstrained	–
Solar wind measurement	Collier *et al.* (1998)	110 ± 290	–	Unconstrained	–
Nebular homogenization to 1 AU	Wasson (1988)	Identical to bulk Earth		0	0
Secularly varying hot solar nebula	Choi and Wasson (1997)	Below TFL†		<0	?
Non-mass-dependent fractionation	Thiemens (1996)	TFL Between ^{16}O Lines		0	~0
Non-mass-dependent fractionation	Nuth *et al.* (1998)	~−40	~−40	~−19	~63
Gas–solid equilibration; R chondrites	Clayton (1993)	>7.6	>6.8	>2.8	>4.1
Two-component gas–solid mixing; Murchison	CM84; this paper	12.3–16.4	7.5–11.4	1.1–2.9	7.7–13.4
Two-component gas–solid mixing; Orgueil	Leshin *et al.* (1997); this paper	12.4	7.8	1.4	8.0
Two-component; solid/gas-enriched 200% in meteorite-forming region	CM84; this paper	33.4	27.9	10.5	37
Two-component; CO → H_2O fractionation at 500° K	CM84; this paper	31.2	19.1	1.1–2.9	30
Two-component; 70% of solids = *GI*	CM84; this paper	16.3–20.4	11.2–15.1	2.7–4.5	13.2–18.8

*This column gives the absolute difference from bulk earth $(\delta^{18}O, \delta^{17}O) = (6, 3)$‰ as described in the text..
†TFL = Terrestrial fractionation line.

1996), or else significantly below the TFL (Nuth *et al.*, 1998). The absolute difference between the CM84 model and the non-mass-dependent fractionation models is ~6‰. A clear distinction between the CM84 model and other predictions can be reached with a measurement of solar composition accurate to several permil.

There are also distinct differences predicted for processes within the mixing model. The last three rows of Table 3 summarize some of the model variations discussed above. The predicted O-isotopic composition of the Sun is very sensitive to the effective solid/gas ratio in the meteorite-forming region. A 200% solid/gas enrichment would be distinctly identifiable in the solar O composition by a displacement of 24‰ farther from bulk Earth along the ^{16}O lines. Similarly, the solar O composition may be sensitive to the temperature at which gaseous conversion of O from CO to H_2O took place upon cooling of the gas. If most of the nebular gas was unconverted at the time of carbonaceous chondrite aqueous alteration, this model would move the predicted solar composition to the right along a mass fractionation line, as indicated by the second line from the bottom of Table 3. The last line shows that the CM84 model is relatively insensitive to whether only some of the solids had an initial ^{16}O-rich composition (*S* in Table 1).

The *Genesis Discovery* mission, due to launch in early 2001, will collect and return samples of solar wind for laboratory analysis. Its highest priority is the measurement of solar wind O-isotopic ratios, including $\delta^{17}O$, to ±1‰. To understand the true solar composition, any differences between solar wind and solar composition must be accounted for. Upper limits on possible mass fractionation due to various processes from the solar convection zone, through the corona, and out to 1 AU are reviewed by Bochsler (1997). Current theory (Von Steiger and Geiss, 1989; Marsch *et al.*, 1995) suggests that there is essentially no isotopic fractionation associated with the first ionization potential fractionation seen between the photosphere and time-averaged solar wind in the abundances of elements heavier than He. There is still a possibility that inefficient Coulomb drag could cause low levels of isotopic fractionation of the time-averaged solar wind relative to the photosphere (Bodmer and Bochsler, 1998, unpubl. data; cf., also Kallenbach *et al.*, 1998). This is an important issue to be tested by *Genesis* independent of the O measurements. Assuming that solar wind isotopic fractionation is nonexistent or small on a permil scale, *Genesis* solar wind O measurements will distinguish between the various solar system O theories. It should provide a baseline by which to compare other solar system materials and should give valuable data on solar system formation. Other future measurements will also be important for validating the O model. For example, if the two-component mixing model is correct, there should be a compositional gradient as one reaches portions of the solar system that were never strongly heated, allowing further tests of the model. For example, do the icy satellites of Jupiter have bulk O compositions intermediate between the inner solar system and, say, comets, which should have primarily sampled the gaseous component?

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APPENDIX

Constraint on Solar Oxygen Isotope Composition Changes Resulting From Early Solar Ionized Outflow

Young stars are generally characterized by strong outflows. A pre-main-sequence outflow might have fractionated the Sun from its initial solid/gas constituent ratio. This is an elemental fractionation that would not directly affect isotope ratios but would cause the ratio R in Table 1 to be incorrect. That ratio is based on the present-day solar elemental composition (*e.g.*, Anders and Grevesse, 1989) by assuming that rock-forming elements entered the solar nebula in oxidized form. If $R_{present\ Sun} < R_{solar\ nebula}$, the solar O composition predicted by the CM84 model using $R_{present\ Sun}$ will be too ^{16}O-poor.

Stellar winds in moderate age stars are characterized by a fractionation based loosely on the first ionization potential (FIP), such that elements with FIP < 10 eV are enriched. Major rock-forming elements, except O, essentially all have low FIP, whereas O and other volatile elements have FIP > 10 eV. The present-day solar wind FIP fractionation factor is ~2 in coronal hole wind and ~4 in low-speed solar wind (*e.g.*, von Steiger, 1995). The present solar-wind mass-loss rate is insignificant. However, the early solar history was most likely characterized by strong winds causing significant mass loss. Although mass loss rates can be estimated for many young stellar objects, the integrated mass loss over the early history of the Sun is difficult to estimate. Mass loss rates tend to decrease as accretion decreases, with outflow/accretion generally estimated at ~0.1. Ionized high-velocity jets are inferred as drivers of cold molecular gas outflows, presumably until this gas has cleared away (see Bally and Stahler, 1997, for review). Bacciotti (1997) estimated the ion/neutral fraction based on observed ionization states in gas along the outflow beam of some Herbig–Haro (HH) objects. The gas in these observations was found to be partially ionized (between 5 and 40%) with the fraction decreasing along the jet. As the cold nebular gas is cleared away from these objects, the ion/neutral fraction increases to near complete ionization by T-Tauri phase and subsequent main sequence stellar winds.

For the solar nebula, the total effect of the FIP fractionation can be estimated using a simple Rayleigh fractionation model:

$$R = R_0 f^{(a-1)} \tag{10}$$

Assuming 10% of the proto-solar material is lost (fraction remaining f = 0.9), and assuming for simplicity that all of the material lost has a FIP fractionation factor of 4.0, $R_{present\ Sun}$ could be as much as 27% lower than R_{Nebula}. When corrected for this effect, the predicted solar $\delta^{18}O$ and $\delta^{17}O$ are each lowered by only ~1.7‰ in the CM84 model. This is very likely an overestimate due to the fact that the gas ejected from the objects discussed above is only partially ionized. The conclusion is that such fractionation has relatively little effect on the predicted solar O composition.

One could envision that the average solar nebula elemental composition was different from the present Sun due to other factors, such as due to a late input to the solar convection zone by infalling planetisimals. However, these factors are also most likely very small as discussed by Laughlin and Adams (1997).